Begin
02-22-2024

Emission and Absorption – Population Rate Equations

More about the Photon Scattering Cross Section

By Definition

$$R_{12} = \frac{|\chi|^2 \beta/2}{\Delta^2+\beta^2} = \sigma(\Delta)\phi = \sigma(\Delta)\frac{\tfrac{1}{2}c\varepsilon_0|E_0|^2}{\hbar\omega}$$

where $|\chi|^2 = |\vec{\mu}_{12}\cdot\vec{\varepsilon}E_0/\hbar|^2 = f\,\dfrac{|\vec{\mu}_{12}|^2|E_0|^2}{\hbar^2}$

and Collision free $\rightarrow$ $\boxed{1 \geq f \geq 1/3}$ $\leftarrow$ Collision broadened

This gives us

$$\boxed{\sigma(\Delta) = f\,\frac{\omega\mu_{12}^2}{\hbar c\varepsilon_0\beta}\,\frac{\beta^2}{\Delta^2+\beta^2} \equiv \sigma(0)\frac{\beta^2}{\Delta^2+\beta^2}}$$

Let $\beta = A_{21}/2$, $\Gamma_1 = \Gamma_2 = 0$ (collision free)

$$\boxed{\sigma(0) = f\,\frac{2\omega\mu_{12}^2}{\hbar c\varepsilon_0 A_{21}} = f\,\frac{4\pi}{\hbar\varepsilon_0\lambda}\,\frac{\mu_{21}^2}{A_{21}}} \quad *$$

The connection between A_{21} and μ_{21}^2 is intuitive, derived rigorously during the QED part of OPTI 544

Here we simply note the result :

$$\boxed{A_{21} = \frac{\mu_{12}^2\omega^3}{3\pi\varepsilon_0\hbar c^3}}$$

Substituting in $*$ we get

$$\sigma(0) = f\,\frac{2\omega}{\hbar c\varepsilon_0}\,\frac{3\pi\varepsilon_0\hbar c^3}{\omega^3} = f\,\frac{3\lambda^2}{2\pi}$$

$$\frac{3\lambda^2}{2\pi} \geq \sigma(0) \geq \frac{\lambda^2}{2\pi}$$

Collision free, polarized light

Collision broadened or un-polarized light

– Remarkably simple result –
easy to remember

Why ?

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$$\boxed{A_{21} = \frac{n_{12}^2\,\omega^3}{3\pi\varepsilon_0\hbar c^3}}$$

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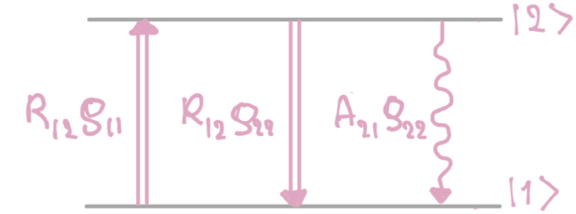
Rate Eq. Model of Absorption

Remember our Physical Picture of absorption/ stim. emission

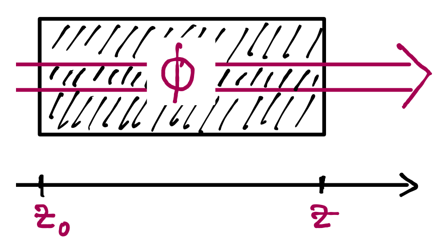


The loss over distance dz is $d\phi = \sigma(\Delta)\phi N(\varrho_{22}-\varrho_{11})$

If $\phi \ll \phi_{sat}$ then $\varrho_{11} \approx 1$ and $\varrho_{22} \approx 0$ and we get

$$d\phi = -N\sigma(\Delta)\phi\,dz \;\Rightarrow\; \frac{d}{dz}\phi = -N\sigma(\Delta)\phi = -\alpha\phi$$

Absorption:



$$\boxed{\begin{aligned}\phi(z) &= e^{-\alpha(z-z_0)}\phi(z_0)\\ I(z) &= e^{-\alpha(z-z_0)}I(z_0)\end{aligned}}$$

Typical $\lambda = 500\,nm$, $\alpha = N \times 10^{-9}\,cm^2\,\frac{\beta^2}{\Delta^2+\beta^2}$

Solid: $N \sim 10^{21}/cm^3$ ⇒ totally opaque @ resonance

Gas: $N \sim 10^{8}/cm^3$ ⇒ transparent @ resonance

Emission and Absorption – Population Rate Equations

Rate Eq. Model of Absorption

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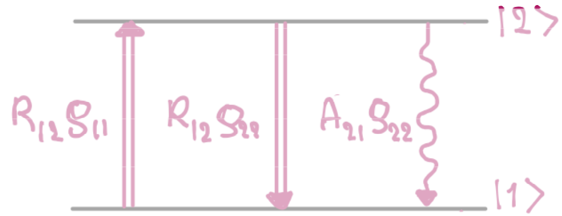

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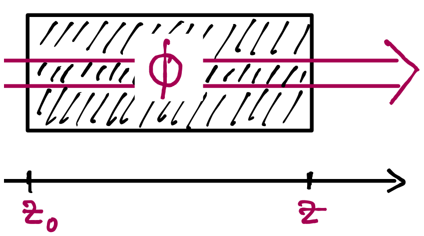
Absorption:



$$\boxed{\begin{aligned} \phi(z) &= e^{-a(z-z_0)}\,\phi(z_0) \\ I(z) &= e^{-a(z-z_0)}\,I(z_0) \end{aligned}}$$

Typical $\lambda = 500\,nm$, $a = N \times 10^{-9}\,cm^2 \dfrac{\beta^2}{\Delta^2+\beta^2}$

Solid: $N \sim 10^{21}/cm^3$ ⇒ totally opaque @ resonance

Gas: $N \sim 10^{8}/cm^3$ ⇒ transparent @ resonance

Note: This is a low saturation result !

High flux ⇒ photon scattering per atom saturates

If $\phi \gg \phi_{sat}$ then $\rho_{22}(\infty) = 1/2$ ⇒ $d\phi = N\dfrac{A_{21}}{2}dz$

$$\phi(z) = \phi(z_0) - \frac{NA_{21}}{2}(z-z_0)$$

At very high flux the loss is relatively insignificant

$$\boxed{\phi(z), \phi(z_0) \gg \frac{NA_{21}}{2}(z-z_0)}$$

– This is referred to as <u>bleaching</u> or <u>hole burning</u>

Emission and Absorption – Population Rate Equations

Blackbody Radiation

This is a standard problem in Optical Physics, which we review only briefly in OPTI 544. See almost any textbook for details, including Milloni & Eberly.

Step (1): We define a Lineshape Function:

$$\boxed{\sigma(\Delta) = \frac{\wp_{12}^2\,\omega_{21}}{2\varepsilon_0 \hbar c}\underbrace{\frac{\delta\nu_{21}/\pi}{(\nu_{21}-\nu)^2+\delta\nu_{21}^2}}_{S(\nu)}\,,\quad \delta\nu_{21}=\beta/2\pi}$$

$S(\nu)$ is sharply peaked compared to the energy density $\rho(\nu)$ of Blackbody Radiation.

$\Rightarrow$ we can approximate $S(\nu)=\delta(\nu_{21}-\nu)$

Step (2): Sub in the expression for transition rates and integrate over all frequencies

$$R_{12}=\int_0^\infty \sigma(\nu)\phi(\nu)\,d\nu = \frac{\wp_{21}^2\,\omega_{21}}{2\varepsilon_0\hbar}\int_0^\infty S(\nu)\frac{\rho(\nu)}{h\nu}\,d\nu$$

$$\Rightarrow\quad R_{12}=\boxed{\frac{\wp_{12}^2}{2\varepsilon_0\hbar^2}}\rho(\nu_{21})=B\rho(\nu_{21})$$

Einstein B coefficient

Step (3): 2-level atom in thermal equilibrium with Blackbody Radiation field

$$\left.\begin{aligned}\frac{\rho_{22}}{\rho_{11}} &= e^{-h\nu_{21}/k_BT}\\ (A_{21}+R_{21})\rho_{22} &= R_{12}\rho_{11}\end{aligned}\right\}\Rightarrow\quad \boxed{\rho(\nu_{21})=\frac{A_{21}/B}{e^{h\nu_{21}/k_BT}-1}}$$

(Detailed Balance)

Step (4): Use prior result for A_{21} to find A_{21}/B which is independent of $\wp_{12}^2$

$$\boxed{\rho(\nu)=\frac{8\pi h\nu^3/c^3}{e^{h\nu/k_BT}-1}}$$

Energy Density of the Blackbody Radiation field

Step (5): We can extend this to find the

* **Total electromagnetic energy density**
* **Total radiated intensity from a black body**

Emission and Absorption – Population Rate Equations

Step (3): 2-level atom in thermal equilibrium with Blackbody Radiation field

$$\left.\begin{aligned} \frac{\varrho_{22}}{\varrho_{11}} &= e^{-h\nu_{21}/k_B T} \\ (A_{21}+R_{21})\varrho_{22} &= R_{12}\varrho_{11} \end{aligned}\right\} \Rightarrow \boxed{\rho(\nu_{21}) = \frac{A_{21}/B}{e^{h\nu_{21}/k_B T}-1}}$$

(Detailed Balance)

Step (4): Use prior result for A_{21} to find A_{21}/B, which is independent of r_{12}^2

$$\boxed{\rho(\nu) = \frac{8\pi h\nu^3/c^3}{e^{h\nu/k_B T}-1}}$$

Energy Density of the Blackbody Radiation field

Step (5): We can extend this to find the

* Total electromagnetic energy density
* Total radiated intensity from a black body

Step (6): Relative importance of spontaneous and stimulated emission

We have $\dfrac{A_{21}}{R_{12}} = \dfrac{A_{21}}{B\rho(\nu)} = e^{h\nu/k_B T}-1 \gg 1$

for $T <$ several $\times$ 1000K

Take the surface of the Sun, $T = 5800\,K$

$$\boxed{\frac{A_{21}}{R_{12}} \sim 140 \ @ \ \lambda_{max} = 500\,nm}$$

End 02-22-2024

Begin
02-27-2024

Vector Model of the 2-Level Atom

We return to the Density Matrix Equations of Motion for a 2-Level atom

$$\dot{\rho}_{11} = -\Gamma_1 \rho_{11} + A_{21}\rho_{22} - \frac{i}{2}(\chi\rho_{12} - \chi^*\rho_{21})$$

$$\dot{\rho}_{22} = -\Gamma_2 \rho_{22} - A_{21}\rho_{22} + \frac{i}{2}(\chi\rho_{12} - \chi^*\rho_{21})$$

$$\dot{\rho}_{12} = (i\Delta - \beta)\rho_{12} + \frac{i\chi^*}{2}(\rho_{22} - \rho_{11}) = \dot{\rho}_{21}^*$$

$$\beta = \frac{1}{T} + \frac{1}{2}(\Gamma_1 + \Gamma_2 + A_{21}), \quad \chi = \vec{\mu}_{21}\cdot\hat{z}\,E_0/\hbar$$

<u>**Note**</u>: In our previous iteration we studied the Rate Equation approximation, which is useful when we are looking for *steady state solutions*

<u>**Here**</u> our goal is different – we seek to recast the Density Matrix formalism in a way that is better suited to understanding and modeling *coherent evolution* and *transient phenomena*. This will also be useful when we study wave and light pulse propagation.

Optical Bloch Equations (OBE's)

Let $\Gamma_1 = \Gamma_2 = 0$ ➡ $\rho_{11} + \rho_{22} = 1, \ \rho_{12} = \rho_{21}^*$

➡ 3 independent, real-valued variables

<u>**Define**</u>	$u = \rho_{21} + \rho_{12}$
Bloch Variables	$v = i(\rho_{21} - \rho_{12})$
	$w = \rho_{22} - \rho_{11}$

Let $\chi = |\chi| e^{-i\varphi}$, substitute in equations for ρ, leaving out relaxation terms $A_{21}, \Gamma_1, \Gamma_2, \beta$

Optical Bloch Equations

$$\dot{u} = -\Delta v - |\chi| \sin\varphi\, w$$

$$\dot{v} = \Delta u + |\chi| \cos\varphi\, w$$

$$\dot{w} = -|\chi|(\cos\varphi\, v - \sin\varphi\, u)$$

Vector Model of the 2-Level Atom

Optical Bloch Equations (OBE's)

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Define:

$$\vec{S} = u\vec{i} + v\vec{j} + w\vec{k}$$
$$\vec{Q} = -|\chi|\cos\varphi\,\vec{i} - |\chi|\sin\varphi\,\vec{j} + \Delta\vec{k}$$

$$\boxed{\dot{\vec{S}} = \vec{Q} \times \vec{S}}$$

Torque Bloch Vector

Equivalent to OBE's !

length conserved

Note: $\frac{d}{dt}(S^2) = 2\vec{S}\cdot\dot{\vec{S}} = 2\vec{S}\cdot(\vec{Q}\times\vec{S}) = 0$

From the definition of the Bloch Variables we get

$$\rho = \frac{1}{2}\begin{pmatrix} 1-w & u+iv \\ u-iv & 1+w \end{pmatrix}$$

and

$$\mathrm{Tr}\,\rho^2 = \frac{1}{2}\left[1 + u^2 + v^2 + w^2\right] = \frac{1}{2}\left[1 + |S|^2\right] \leq 1$$

$$\Rightarrow \boxed{|S|^2 \leq 1}$$

Vector Model of the 2-Level Atom

Define:

$$\boxed{\begin{aligned}\vec{S} &= u\vec{i} + v\vec{j} + w\vec{k} \\ \vec{Q} &= -|X|\cos\varphi\,\vec{i} - |X|\sin\varphi\,\vec{j} + \Delta\vec{k}\end{aligned}}$$

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$$\Rightarrow \boxed{|S|^2 \leq 1}$$

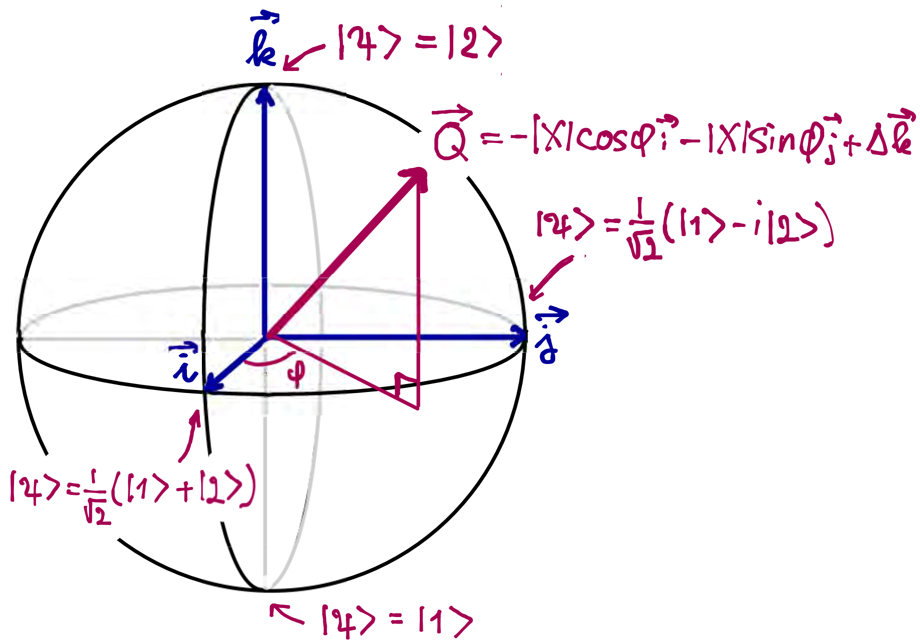
Clearly, $|S^2| = 1 \Rightarrow \mathrm{Tr}\,\rho^2 = 1 \Rightarrow$ pure state

States w/ $|\vec{S}| < 1 \Rightarrow \mathrm{Tr}\,\rho^2 < 1 \Rightarrow$ mixed state

$$|\vec{S}| = 0 \Rightarrow \mathrm{Tr}\,\rho^2 = 1/2 \Rightarrow \rho = \begin{pmatrix} 1/2 & 0 \\ 0 & 1/2 \end{pmatrix}$$

maximally mixed

__Note:__ The above suggests a physical state can be represented by a vector $\vec{S}$, whose tip lies on the surface of (pure) or inside (mixed) a sphere of unit radius, and whose length is conserved under Schrödinger evolution. This is the __Bloch Sphere__.



Vector Model of the 2-Level Atom

Clearly, $|S^2|=1 \Rightarrow \mathrm{Tr}\rho^2=1 \Rightarrow$ pure state

States w/ $|\vec{S}|<1 \Rightarrow \mathrm{Tr}\rho^2<1 \Rightarrow$ mixed state

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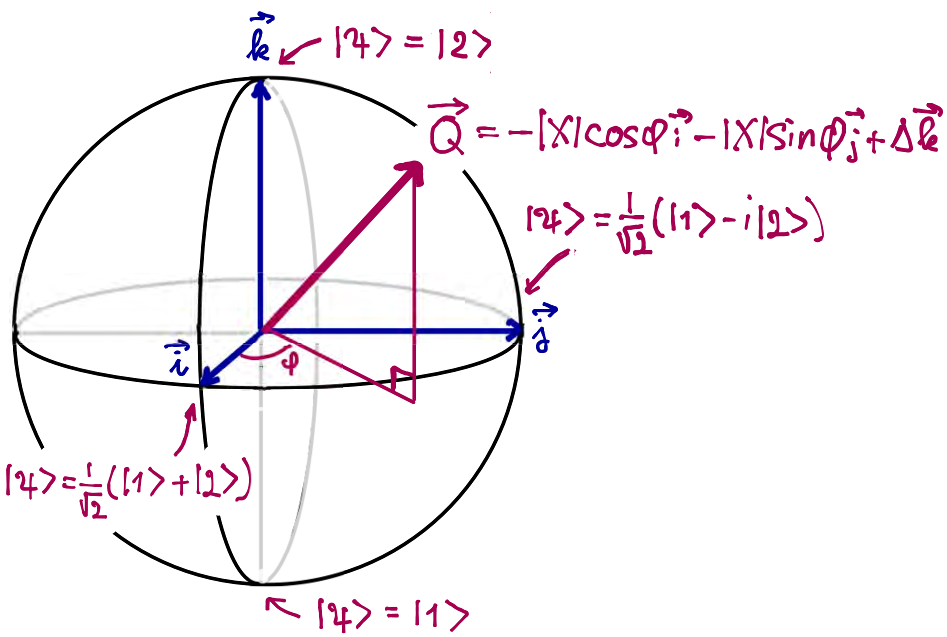

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(∗) <u>Only</u> if the 2-level system is a physical spin-1/2 particle does $\vec{S}$ correspond to an angular momentum vector that lives in physical space. In general, $\vec{S}$ is what we call a *pseudo-spin*, not an actual physical spin.

Physical Interpretation of the Bloch Variables

We have $\langle\vec{\mu}\rangle = \mathrm{Tr}(\rho\vec{\mu}) = \rho_{12}\vec{\mu}_{21} + \rho_{21}\vec{\mu}_{12}$

where
$$\left.\begin{aligned} \rho_{12} &= \tfrac{1}{2}(u+iv)e^{i\omega t} \\ \rho_{21} &= \tfrac{1}{2}(u-iv)e^{-i\omega t} \end{aligned}\right\} \text{fast variables}$$

$$\vec{E} = \mathrm{Re}[\vec{\varepsilon}E_0 e^{-i\omega t}] \quad \text{driving field}$$

It follows that

$$\boxed{\begin{aligned} \langle\vec{\mu}\rangle &= \tfrac{1}{2}(u+iv)e^{i\omega t}\vec{\mu}_{21} + \tfrac{1}{2}(u-iv)e^{-i\omega t}\vec{\mu}_{12} \\ &= u\,\mathrm{Re}[\vec{\mu}_{21}e^{-i\omega t}] + v\,\mathrm{Im}[\vec{\mu}_{12}e^{-i\omega t}] \end{aligned}}$$

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Thus

u is the component of $\langle\hat{\vec{\mu}}\rangle$ in-phase w/ $\vec{E}$

v is the component of $\langle\hat{\vec{\mu}}\rangle$ in-quadrature w/ $\vec{E}$

Lastly, $w = \rho_{22} - \rho_{11}$ is the population inversion.

Solution of the OBE's

Let $\Delta = 0$ and χ real and positive ⇨ $\begin{cases}\vec{Q} = -|\chi|\vec{i}\\ \varphi = 0\end{cases}$

$$\boxed{\begin{aligned}\vec{S} &= u\vec{i} + v\vec{j} + w\vec{k}\\ \vec{Q} &= -|\chi|\cos\varphi\,\vec{i} - |\chi|\sin\varphi\,\vec{j} + \Delta\vec{k}\end{aligned}}$$

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choose global phase so u(0) = 0

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Solution

Rabi Oscillations

$$\boxed{\begin{aligned} u &= 0 \\ v &= -\sin\theta\,, \quad \theta = \chi t \\ w &= -\cos\theta \end{aligned}}$$

Vector Model of the 2-Level Atom

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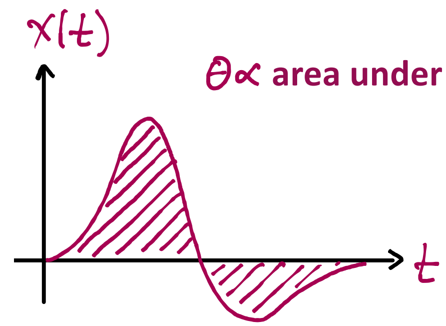
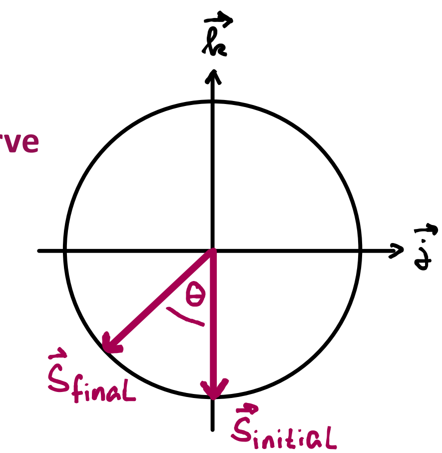
What if $\chi = \chi(t)$? We now have solutions

$$\boxed{\begin{aligned} u &= 0 \\ v &= -\sin\theta\ , \quad \theta = \int_0^t \chi(t')\,dt' \\ w &= -\cos\theta \end{aligned}}$$

Pulse Area Theorem!

This is a very important result ⇒ We can deal with pulses !





Note: $\chi(t) = \overbrace{\vec{p}_{21}\cdot\vec{\varepsilon}}^{\mu}\,E(t)/\hbar \equiv \mu\,\mathcal{E}(t)/\hbar$

complex amplitude

Cannot remain real if the complex phase of $\mathcal{E}(t)$ is changing with time

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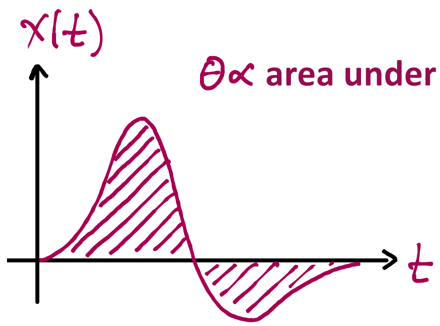
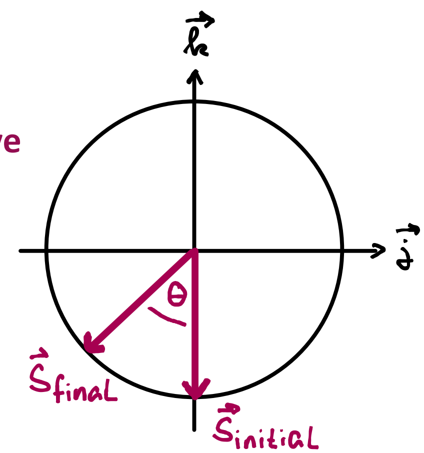
Pulse Area Theorem!

This is a very important result ⇨ We can deal with pulses!





Note: $\chi(t) = \overbrace{\vec{\mu}_{21}\cdot\vec{\varepsilon}}^{\mu}\, E(t)/\hbar \equiv \mu\,\mathcal{E}(t)/\hbar$

complex amplitude

Cannot remain real if the complex phase of $\mathcal{E}(t)$ is changing with time

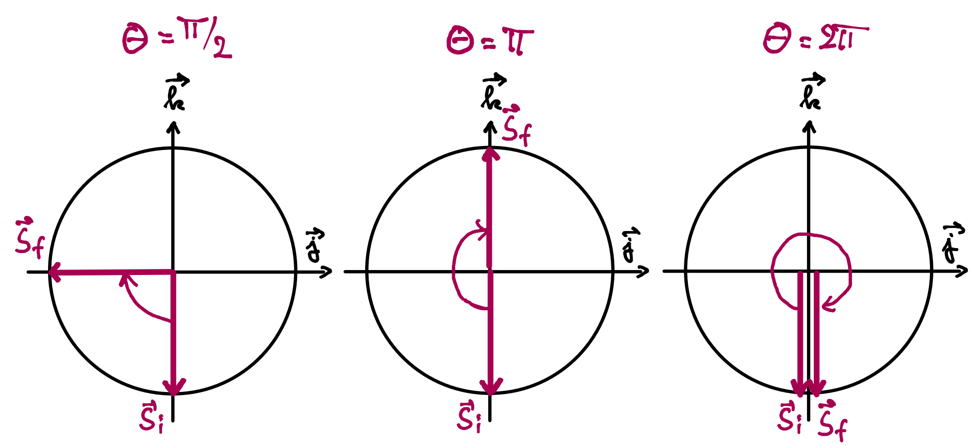
Different phase ⇨ different axis of rotation

The Pulse Area Theorem is only valid if the <u>direction</u> of the torque vector $\vec{Q}$ is constant.

<u>Note</u>: The RWA is valid only in the Slowly Varying Envelope Approx. $\Big\}\ \frac{d\mathcal{E}}{dt} \ll \omega$

This may not hold for modern Ultrafast Lasers !

Some examples of θ-pulses: (quantum gates)



Vector Model of the 2-Level Atom

Different phase → different axis of rotation

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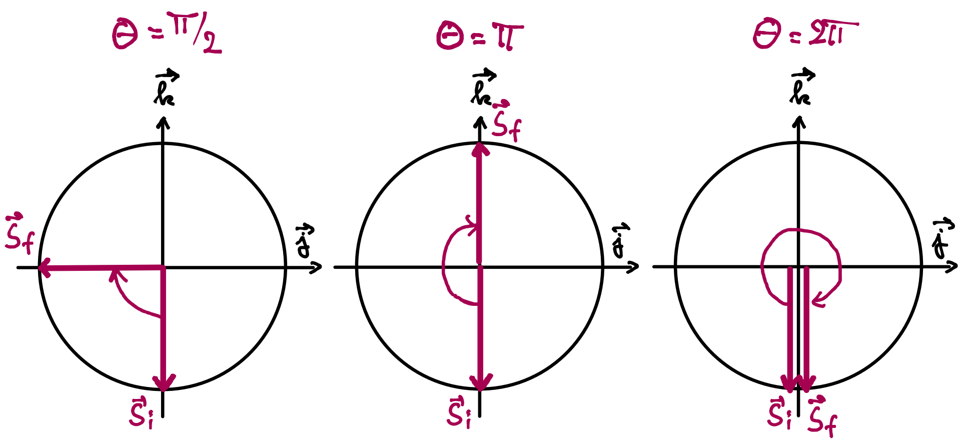
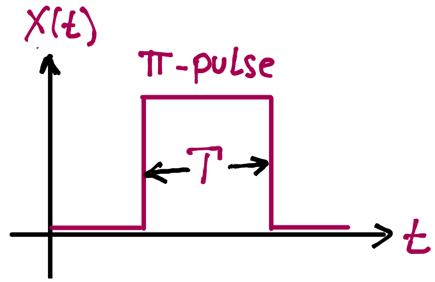
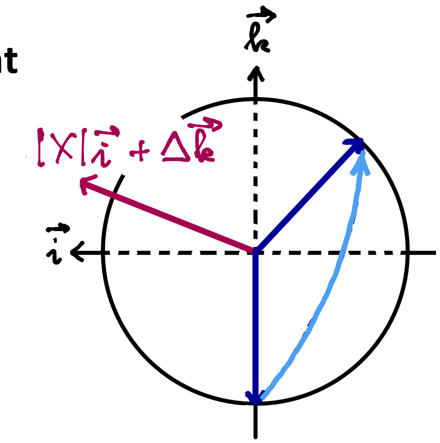


Ramsey Method of Separated Oscillatory Fields

(The Ramsey “trick”, 1989 Nobel in Physics)

Single pulse measurement of ω_{21} in Atomic Clock:





Idea is to measure population of $|2\rangle$ as function of $\Delta = \omega_{21} - \omega$ which is maximized for $\omega = \omega_{21}$.

The frequency resolution is $\delta\omega \propto 1/T$, so very long pulses are required. The atom is perturbed by interaction with the light during the entire interrogation and problems can occur due to phase or amplitude noise on the light field. This is not good since the clock is supposed to link to the transition frequency of an unperturbed atom.

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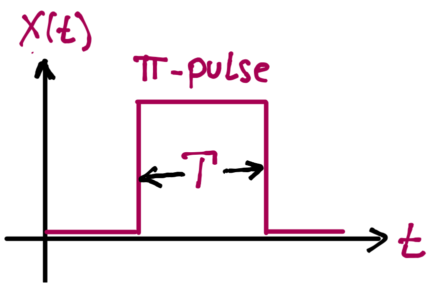

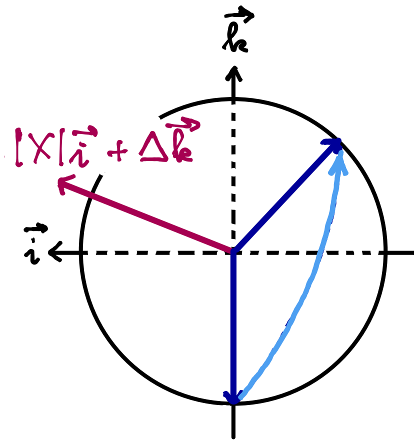

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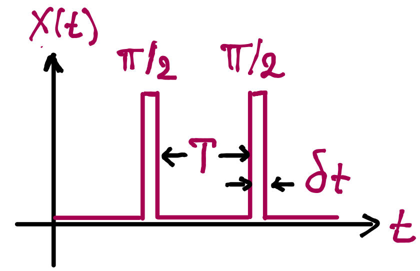
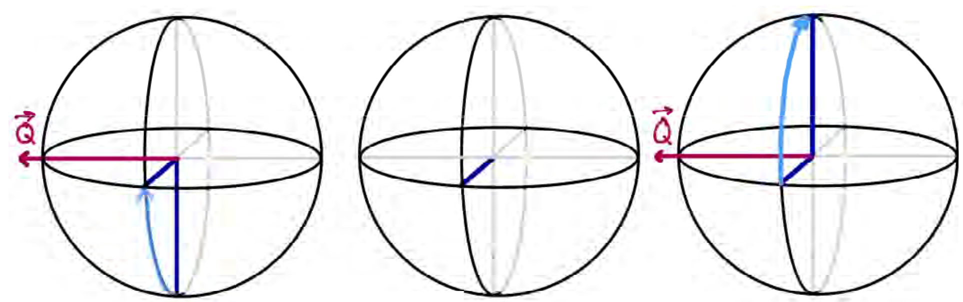
Ramsey's two-pulse strategy:



Sequence of 2 short, intense $\pi/2$ pulses, separated by a long free evolution period, So that $T \gg \delta t$

During pulses $|\chi| \gg \Delta$ $\Rightarrow$ $\vec{Q} = |\chi|\vec{i} + \Delta\vec{k} \sim |\chi|\vec{i}$

Case $\Delta = 0$, path on the Bloch sphere



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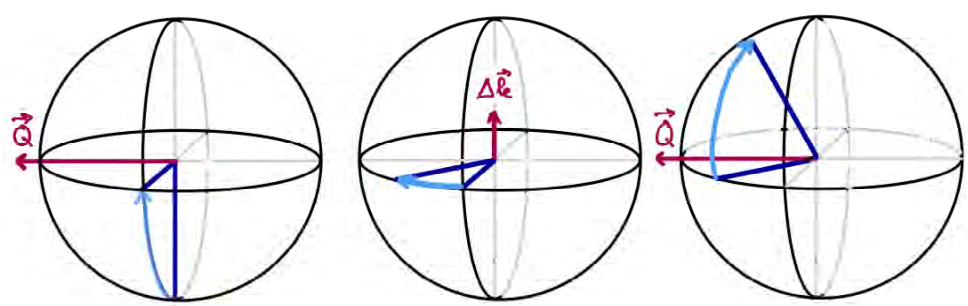

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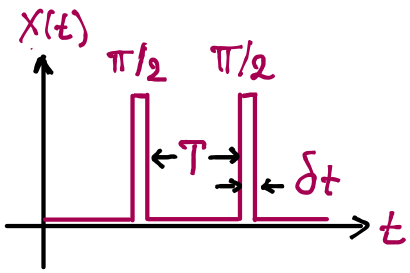

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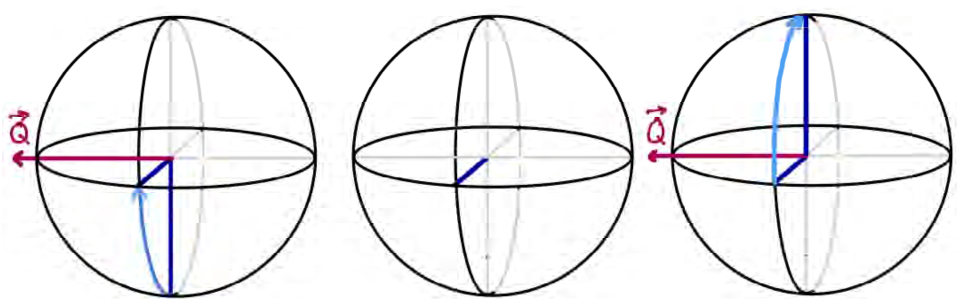

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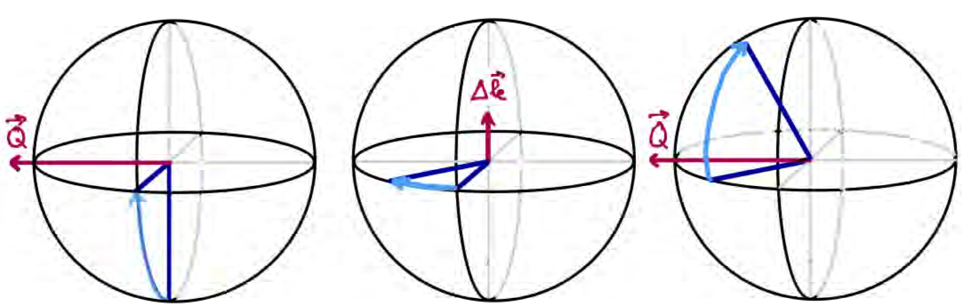

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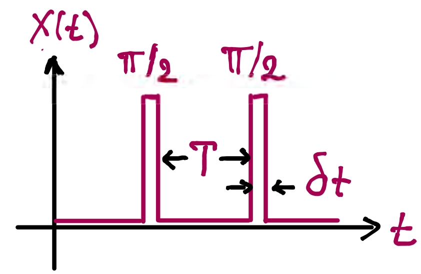

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Again, if we measure the population of $|2\rangle$ as a function of $\Delta = \omega_{21} - \omega$, a maximum is found when $\omega = \omega_{21}$. However, the resolution is now $\delta\omega \propto 1/T$ where T is the time *between* pulses. This is an enormous advantage for atomic clocks and other forms of precision metrology.

Vector Model of the 2-Level Atom

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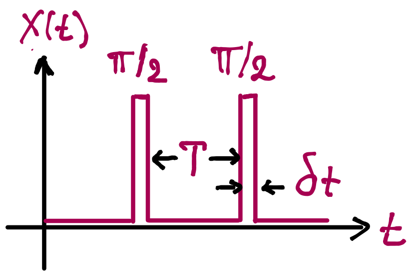

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During pulses $|X| \gg \Delta$ ➡ $\vec{Q} = |X|\vec{i} + \Delta\vec{k} \sim |X|\vec{i}$

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